

# Frequency-Doublings with Symmetry-Broken Surface-State Electrons

Miao Zhang,<sup>1</sup> W. Z. Jia,<sup>1</sup> and L. F. Wei<sup>\*1,2</sup>

<sup>1</sup>Quantum Optoelectronics Laboratory, School of Physics and Technology,  
Southwest Jiaotong University, Chengdu 610031, China

<sup>2</sup>State Key Laboratory of Optoelectronic Materials and Technologies,  
School of Physics and Engineering, Sun Yat-sen University, Guangzhou 510275, China  
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Frequency-doubling is one of important ways to obtain high-frequency electromagnetic radiations, and is usually realized by using various nonlinear optical effects in crystals. Here, we propose an alternative approach to implement such a process with the surface-state electrons on the liquid Helium. Due to the symmetry-broken eigenstates of electrons, we show that the like-Rabi oscillations between two levels of the surface-state electrons could be realized *beyond the usual resonant drivings*. Consequently, an electromagnetic field with the doubled frequency of the applied driving is remarkably radiated. Importantly, such a frequency-doubling effect could be utilized to generate high-frequency microwave radiations, up to Terahertz (THz) ones.

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**Introduction.**— An electron (with mass  $m_e$  and charge  $e$ ) near the surface of liquid Helium is weakly attracted by its dielectric image potential  $V(z) = -\Lambda e^2/z$ . Where  $z$  is the distance above liquid Helium surface, and  $\Lambda = (\epsilon - 1)/4(\epsilon + 1)$  with  $\epsilon$  being dielectric constant of the liquid Helium [1]. Due to the Pauli exclusion principle, there is an barrier about 1 eV for preventing the electron penetrating into the liquid Helium. Together with such a hard wall at  $z = 0$ , the electron is resulted in a one-dimensional (1D) hydrogenlike spectrum  $E_n = -R/n^2$  of motion normal to the Helium surface, with Rydberg energy  $R = \Lambda^2 e^4 m_e / (2\hbar^2) \approx 0.17$  THz and effective Bohr radius  $r_B = \hbar^2 / (m_e e^2 \Lambda) \approx 76$  Å. These Rydberg levels were first observed by Grimes *et al.* [2, 3] by measuring the resonant frequencies of the transitions between the ground and excited states, and recently be proposed to implement quantum information processings [4, 5].

In the past decades, many interests were paid on the linewidths of the above Rydberg states, see, e.g., [6, 7]. Since the liquid Helium is very cold (e.g.,  $T \approx$  mK), the vapor of Helium atoms above the surface is negligible and thus the noises are mainly originated from the thermally-excited surface waves (i.e., the so-called ripplons [8–10]). However, the amplitudes ( $\approx 0.2$  Å) of the ripplons are still significantly smaller than the effective Bohr radius (due to the very-weak image potential), and thus the Rydberg states of the electrons have the relatively-long lifetimes (and thus significantly small linewidths [6]). Recently, strong interests have been paid to the quantum manipulations of the surface-state electrons on the liquid Helium. This is due to a remarkable proposal suggested by Platzman and Dykman [4], who suggested that the surface-state electrons could be utilized to implement the quantum information processings. Up to now, several approaches have been proposed to perform the coherent operations of surface-state electrons in the single-electron regime, see, e.g., [11–15].

Here, we show that the surface-state electrons on the liquid Helium could be utilized to realize the frequency-doubling

at high-frequency microwave regime. Usually, the optical frequency-doubling, called also second harmonic generation, is obtained by using various nonlinear interactions between the lights and natural-atoms. For example, a pump light propagating through a crystal with second-order susceptibility  $\chi^{(2)}$  could generate a nonlinear polarization response, which radiates an electromagnetic field with the doubled frequency of the pump light. This phenomenon was first observed by Franken *et al* [16], and has been applied to numerous areas in both science and engineering [17]. Different from the usual schemes to implement the frequency-doubling, we realize this process *by using the unusual Stark effects related to the parity symmetry-breakings of the electronic surface-states*. Our idea is based on the fact: the surface-state electrons allows an unusual dipole moment which is related to the nonzero average distances between the Rydberg states, i.e.,  $\mathcal{Z} = \langle m|z|m \rangle - \langle n|z|n \rangle \neq 0$  [3, 5, 18]. Basically, under the resonant drivings the usual transition  $\langle n|z|m \rangle$  is dominant and the effects related to  $\mathcal{Z}$  are negligible. Alternatively, under the certain large-detuning drivings, the effects related to the  $\mathcal{Z}$  become important, and consequently the desirable frequency-doublings could be realized in a new way.

**Like-Rabi oscillations beyond the resonant drivings.**—We consider applying a weak microwave field  $\mathcal{E} = \mathcal{E} \cos(kr + \omega_l t + \phi)$  to a single surface-state electron, where  $\mathcal{E}$ ,  $k$ ,  $\omega_l$ , and  $\phi$  are its amplitude (in  $z$  direction), wave vector, frequency, and initial phase, respectively. For simplicity, we consider only two levels (e.g., the ground state  $|1\rangle$  and the first excited state  $|2\rangle$ ) of the surface-state electron. Also, the electron is assumed to move within a small region whose size is much smaller than the wavelength of the applied microwave. Therefore, under the usual dipole approximation ( $kr \approx 0$ ) the Hamiltonian describing the driven two-level electron reads

$$\hat{H} = E_1|1\rangle\langle 1| + E_2|2\rangle\langle 2| - e\hat{z}\mathcal{E} \cos(\omega_l t + \phi). \quad (1)$$

By using the completeness relation  $|1\rangle\langle 1| + |2\rangle\langle 2| = \hat{1}$ , one can write  $\hat{z}$  as

$$\hat{z} = z_{11}|1\rangle\langle 1| + z_{22}|2\rangle\langle 2| + z_{12}|1\rangle\langle 2| + z_{21}|2\rangle\langle 1|, \quad (2)$$

with the electric-dipole matrix elements  $z_{12} = \langle 1|\hat{z}|2\rangle = \langle 2|\hat{z}|1\rangle = z_{21}$ . Due to the asymmetry eigenstates  $|1\rangle$  and

\*weilianf@mail.sysu.edu.cn; weilianfu@gmail.com

$|2\rangle$ , here  $z_{11} = \langle 1|\hat{z}|1\rangle$  and  $z_{22} = \langle 2|\hat{z}|2\rangle \neq 0$ . This is very different from the case for the natural atoms wherein  $z_{11} = z_{22} = 0$ . By using Eq. (2) we can rewrite Hamiltonian (1) as

$$\hat{H} = \frac{\hbar\omega_e}{2}\hat{\sigma}_z - \frac{1}{2} \left[ \hbar\tilde{\Omega}\hat{\sigma}_z + \hbar\Omega_R(\hat{\sigma}_{12} + \hat{\sigma}_{21}) \right] \cos(\omega_l t + \phi), \quad (3)$$

and further

$$\begin{aligned} \hat{H}_I(t) = & -\hbar\tilde{\Omega}\hat{\sigma}_z (e^{i\omega_l t + i\phi} + e^{-i\omega_l t - i\phi}) \\ & -\hbar\Omega_R \left[ e^{-i\Delta t + i\phi}\hat{\sigma}_{12} + e^{i\Delta t - i\phi}\hat{\sigma}_{21} \right. \\ & \left. + e^{-i(\omega_e + \omega_l)t - i\phi}\hat{\sigma}_{12} + e^{i(\omega_e + \omega_l)t + i\phi}\hat{\sigma}_{21} \right], \end{aligned} \quad (4)$$

in the interaction picture defined by  $\hat{R} = \exp(-it\omega_e\hat{\sigma}_z/2)$ . Above,  $\hat{\sigma}_z = |2\rangle\langle 2| - |1\rangle\langle 1|$  is the usual Pauli operator with the transition frequency  $\omega_e = (E_2 - E_1)/\hbar$ ,  $\hat{\sigma}_{ij} = |i\rangle\langle j|$  is the projection operator ( $i, j = 1, 2$ ),  $\Delta = \omega_e - \omega_l$  is the detuning between the microwave and electron,  $\Omega_R = z_{12}e\mathcal{E}/(2\hbar)$  is the usual Rabi frequency, and finally  $\tilde{\Omega} = (z_{22} - z_{11})e\mathcal{E}/(4\hbar) \neq 0$  due to the symmetry-breaking in the states  $|1\rangle$  and  $|2\rangle$ . Typically,  $(\Omega_R, \tilde{\Omega})/(\omega_l, \omega_e) \ll 1$  within the weak drivings regime.

Under the weak drivings near the resonant point, i.e.,  $\Delta \ll (\omega_l, \omega_e)$ , the evolution ruled by the Hamiltonian (4) can be approximately expressed as [19]

$$\begin{aligned} \hat{U}(t) = & \hat{T} \exp \left[ \int_0^t \hat{H}_I(t) dt \right] \\ = & 1 + \left( \frac{-i}{\hbar} \right) \int_0^t \hat{H}_I(t_1) dt_1 + \dots \\ \approx & \hat{T} \exp \left[ \int_0^t \hat{H}_R(t) dt \right], \end{aligned} \quad (5)$$

with the Dyson-series operator  $\hat{T}$  and the effective Hamiltonian  $\hat{H}_R(t) = -\hbar\Omega_R(e^{-i\Delta t + i\phi}\hat{\sigma}_{12} + e^{i\Delta t - i\phi}\hat{\sigma}_{21})$ . In the rotating-framework defined by the unitary operator  $\hat{R}_R = \exp(it\Delta\hat{\sigma}_z/2)$ , this effective Hamiltonian reads

$$\hat{H}_R = \frac{\hbar\Delta}{2}\hat{\sigma}_z - \hbar\Omega_R(e^{i\phi}\hat{\sigma}_{12} + e^{-i\phi}\hat{\sigma}_{21}). \quad (6)$$

Certainly, when  $\Delta = 0$ , the Hamiltonian (6) describes the standard Rabi oscillation with the frequency  $\Omega_R$ . In Eq. (5), the terms related to  $\xi = (\Omega_R, \tilde{\Omega})/(\omega_l, \omega_e)$  are neglected, since  $\xi \ll 1$  under the weak drivings. In fact, this approximation is nothing but the usual rotating-wave approximation (RWA) [19]. Indeed, under the resonant excitation, i.e.,  $\Delta \ll (\omega_l, \omega_e)$ , the contribution from  $\hbar\tilde{\Omega}\hat{\sigma}_z$  (due to the symmetry-breaking of the present surface-state electrons) should be negligible, due to its small probability proportional to  $\xi$ .

More interestingly, under the large-detuning driving, e.g.,  $\Delta = \omega_l - \delta$  (with a small adjustment  $\delta \sim 0$ ), the effects related to the Stark term  $\hbar\tilde{\Omega}\hat{\sigma}_z$  become significant. This can

be seen from the following new evolution operator

$$\begin{aligned} \hat{U}(t) = & \hat{T} \exp \left[ \int_0^t \hat{H}_I(t) dt \right] \\ = & 1 + \left( \frac{-i}{\hbar} \right) \int_0^t \hat{H}_I(t_1) dt_1 \\ & + \left( \frac{-i}{\hbar} \right)^2 \int_0^t \hat{H}_I(t_2) \int_0^t \hat{H}_I(t_1) dt_1 dt_2 + \dots \\ \approx & \hat{T} \exp \left[ \int_0^t \hat{H}_L(t) dt \right], \end{aligned} \quad (7)$$

with the so-called second-order effective Hamiltonian [20]  $\hat{H}_L(t) = \hbar\nu\hat{\sigma}_z/2 - \hbar\Omega_L(e^{i\delta t + i2\phi}\hat{\sigma}_{12} + e^{-i\delta t - i2\phi}\hat{\sigma}_{21})$ , which can be further simplified as

$$\hat{H}_L = \frac{\hbar\Delta'}{2}\hat{\sigma}_z - \hbar\Omega_L(e^{i2\phi}\hat{\sigma}_{12} + e^{-i2\phi}\hat{\sigma}_{21}) \quad (8)$$

in the rotating framework defined by  $\hat{R}_L = \exp(-it\delta\hat{\sigma}_z/2)$ . Above, the value of  $\delta = 2\omega_l - \omega_e$  is controllable by the selected frequency  $\omega_l$  of the applied microwave,  $\Delta' = \nu - \delta$  with  $\nu = 4\Omega_R^2[1/(\omega_e - \delta) + 1/(3\omega_e + \delta)]$ . While, due to the weak driving ( $\Omega_R, \tilde{\Omega} \ll (\omega_l, \omega_e)$ ), the present Rabi frequency  $\Omega_L = 4\Omega_R\tilde{\Omega}\omega_e/(\omega_e^2 - \delta^2)$  is smaller than the previous one  $\Omega_R$  for the resonant excitation. Also, all the effects related to the small quantity  $\xi = (\Omega_R, \tilde{\Omega})/(\omega_l, \omega_e)$  were neglected, but the terms containing  $\Omega_R\tilde{\Omega}/(\omega_l, \omega_e)$  were retained. Note that, under the above large-detuning driving, the first-order expansion term  $\hat{U}^{(1)}(t) = (-i/\hbar) \int_0^t \hat{H}_I(t_1) dt_1$  practically does not contribute to the time evolution, due to its small probability:  $P^{(1)}(t) \sim \xi$ .

Remarkably, the above large-detuning driving can also induce the oscillating occupancies of the surface-state energies, i.e., like-Rabi oscillation. In fact, due to  $\delta \ll (\omega_l, \omega_e)$  we have the following approximated expansion

$$\begin{aligned} \Delta' = & \frac{4\Omega_R^2}{\omega_e} \sum_{n=0}^{\infty} \left[ 1 + \frac{1}{3} \left( \frac{-1}{3} \right)^n \right] \left( \frac{\delta}{\omega_e} \right)^n - \delta \\ \approx & \frac{16\Omega_R^2}{3\omega_e} + \left( \frac{32\Omega_R^2}{9\omega_e^2} - 1 \right) \delta. \end{aligned} \quad (9)$$

Obviously, if the “new detuning”  $\Delta' = 0$ , i.e.,  $\delta = 48\omega_e\Omega_R^2/(9\omega_e^2 - 32\Omega_R^2)$  and  $\omega_l = \omega_e/2 + 24\omega_e\Omega_R^2/(9\omega_e^2 - 32\Omega_R^2)$ , the Hamiltonian (8) defines to a like-Rabi oscillation with the frequency  $\Omega_L$ .

The above analytical analysis can be confirmed by the numerical methods. We consider the typical parameters [3, 5]: the transition frequency  $\omega_e \approx 220$  GHz, matrix elements  $z_{12} \approx 0.5 r_B$ ,  $z_{22} - z_{11} \approx 2.3 r_B$ , and the strength  $\mathcal{E} = 15$  V/cm of the applied microwave. Consequently, we have  $\Omega_R \approx 4.3$  GHz,  $\tilde{\Omega} \approx 10$  GHz,  $\xi = (\Omega_R, \tilde{\Omega})/(\omega_l, \omega_e) < 1/10$ , and thus  $\Omega_L \approx 0.8$  GHz for  $\Delta' = 0$ . Fig. 1(a) shows the oscillations of the occupancies  $\rho_{22}$  in the excited state  $|2\rangle$  versus time. This figure includes the analytical results from effective Hamiltonian (8) (with the RWA,  $\Delta' = 0$ , and  $\Omega_L \approx 0.8$ ) and the numerical ones from the original Hamiltonian (3) without any approximation. One can see that the performed RWA, i.e., neglecting the terms related to  $\xi$ , works

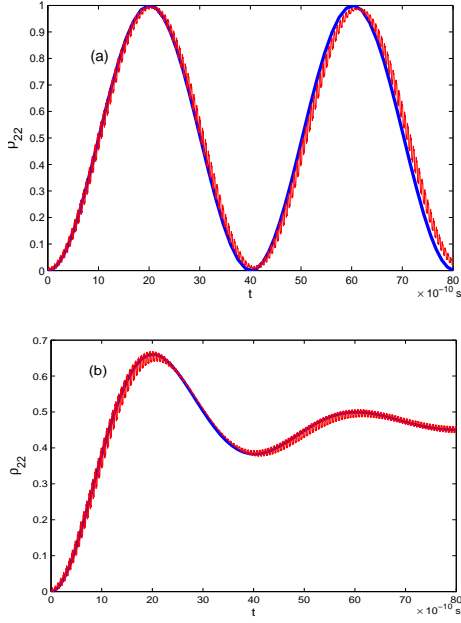


FIG. 1: Oscillating occupancy  $\rho_{22}$  in the excited state  $|2\rangle$ : (a) without dissipation, (b) with the induced dissipations  $\Gamma = \gamma = g/10$ . Here, the blue and red lines correspond to the analytical solution from the effective Hamiltonian (8) and the numerical one from the original Hamiltonian (3), respectively.

very well. In principle, if the strength  $\mathcal{E}$  of the applied microwave is selected significantly small, then  $\xi$  is sufficiently small, and thus our effective Hamiltonian (8) is more exact. Worth of note, for the natural atoms (where  $\tilde{\Omega} = 0$ ), the above like-Rabi oscillation vanishes, which means that the natural atoms can not be effectively driven by the large-detuning driving. This can be easily verified numerically, e.g.,  $\rho_{22} < 1.2\%$ .

*Frequency-doubling radiations.*—We now consider the steady-state radiations due to the above oscillations of occupancies by taking into account the practically-existing dissipations. For this case the dynamics of the driven surface-state electrons is described by the master equation [19]

$$\begin{aligned} \frac{d\hat{\rho}}{dt} = & \frac{-i}{\hbar} [\hat{H}_L, \hat{\rho}] \\ & + \frac{\Gamma}{2} (2\hat{\sigma}_{12}\hat{\rho}\hat{\sigma}_{21} - \hat{\sigma}_{21}\hat{\sigma}_{12}\hat{\rho} - \hat{\rho}\hat{\sigma}_{21}\hat{\sigma}_{12}) \\ & + \frac{\gamma}{2} (2\hat{\sigma}_{22}\hat{\rho}\hat{\sigma}_{22} - \hat{\sigma}_{22}\hat{\rho} - \hat{\rho}\hat{\sigma}_{22}), \end{aligned} \quad (10)$$

with  $\Gamma$  and  $\gamma$  being the decay and dephasing rates, respectively. Above,  $\hat{\rho} = \sum_{i,j=1}^2 \rho_{ij} |i\rangle\langle j|$  is the density operator of the two-level quantum system, and the matrix elements  $\{\rho_{ij}\}$  obey the normalized and hermitian conditions:  $\sum_{i=1}^2 \rho_{ii} = 1$  and  $\rho_{ij} = \rho_{ji}^*$ , respectively. In the two-level atomic representation, the above master equation takes the following matrix

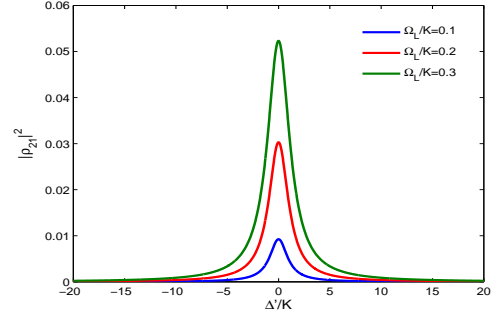


FIG. 2: Within the weak interaction regime  $\Omega_L/K = 0.1, 0.2, 0.3$ , and  $\Gamma = \gamma$ , relative intensity  $|\rho_{21}|^2$  of the frequency-doubling radiation versus the detuning parameter  $\Delta'/K$ .

forms:

$$\begin{cases} \dot{\rho}_{11} = i\Omega_L(\rho_{21}e^{i\phi} - \rho_{12}e^{-i\phi}) + \Gamma\rho_{22}, \\ \dot{\rho}_{22} = i\Omega_L(\rho_{12}e^{-i\phi} - \rho_{21}e^{i\phi}) - \Gamma\rho_{22}, \\ \dot{\rho}_{21} = i(\rho_{11}\Omega_L e^{-i\phi} - \rho_{22}\Omega_L e^{-i\phi} - \Delta\rho_{21}) \\ \quad - \frac{\Gamma}{2}\rho_{21} - \frac{\gamma}{2}\rho_{21}. \end{cases} \quad (11)$$

Certainly, due to the resistance from the surroundings, the amplitudes of oscillating  $\rho_{ii}$  decrease with the time evolving (see, e.g., Fig. 1(b)), and  $\dot{\rho}_{ii} \rightarrow 0$  when  $t \rightarrow \infty$ . Specially, under the steady-state condition:  $\dot{\rho}_{ij} = 0$ , we have

$$\begin{cases} \rho_{22} = \frac{2\Omega_L^2\mathcal{K}}{\Gamma(\mathcal{K}^2 + \Delta^2) + 4\Omega_L^2\mathcal{K}}, \\ \rho_{21} = \frac{\Omega_L e^{-i2\phi}(\rho_{11} - \rho_{22})(i\mathcal{K} + \Delta)}{\mathcal{K}^2 + \Delta^2}, \end{cases} \quad (12)$$

with  $\mathcal{K} = (\Gamma + \gamma)/2$ .

Immediately, we can compute the polarization  $\mathcal{P} = \langle ez \rangle = \text{Tr}(e\hat{\rho}\hat{z}) = e \sum_{i,j=1}^2 \rho_{ij} z_{ji}$  for single surface-state electrons. Note that the Eqs. (11) and (12) are obtained in the rotating-framework with frequency  $\omega_e + \delta = 2\omega_l$ . Transforming back to the Schrödinger picture, the polarization of the electrons reads

$$P = e (\rho_{21}z_{12}e^{-i2\omega_l t} + \rho_{12}z_{21}e^{i2\omega_l t} + \rho_{11}z_{11} + \rho_{22}z_{22}). \quad (13)$$

This indicates that the surface-state electron has the steady  $2\omega_l$ -response for the applied incident field  $\mathcal{E} = \mathcal{E} \cos(\omega_l t)$ . Such a  $2\omega_l$ -response acts further as an effective source of new radiation  $\mathcal{E}_2$  with doubling-frequency  $2\omega_l$ .

By discarding the static polarizations  $\rho_{11}z_{11}$  and  $\rho_{22}z_{22}$  (which practically do not contribute to the generations of the radiations), we can write the polarization (13) in a simple form

$$\mathcal{P} = \mathcal{A} \cos(2\omega_l t - \theta), \quad (14)$$

with the amplitude  $\mathcal{A} = e z_{12} \sqrt{\rho_{21}\rho_{12}}$  and phase  $\theta = \arctan(\text{Im}\rho_{21}/\text{Re}\rho_{21}) + 2\phi$ . It has been well known that the radiation field of an electric dipole is determined by its

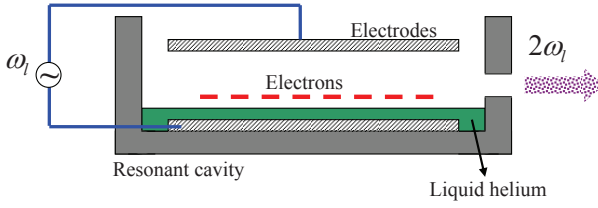


FIG. 3: A sketch of the experimental setup to obtain the frequency-doubling radiations.

polarization, i.e.,  $\mathcal{E}_2 \propto |\ddot{\mathcal{P}}|$ . Thus, the intensity  $I_2$  of the generated frequency-doubling wave here is proportional to  $|\rho_{21}|^2$ . Within the usual weak-interaction regime [19], i.e.,  $\Omega_L/KT \ll 1$ , we have

$$|\rho_{21}|^2 \approx \frac{(\Omega_L/K)^2}{1 + (\Delta'/K)^2 + 8(\Omega_L^2/KT)}. \quad (15)$$

Obviously, the maximum radiating intensity appears at  $\Delta' = 0$  (see, Fig. 2), and it could be increased significantly by increasing the Rabi frequency  $\Omega_L$ , via enhancing the strengths of the driven field  $\mathcal{E}$ .

**Discussions and Conclusions.**— The frequency-doubling radiations proposed above could be directly realized by using the experimentally-existing setups, e.g., shown in Fig. 3 [2–4]. Where, a number of electrons are confined on the liquid Helium surface by the designed static-electric fields [11, 12], and driven by a spatial-uniform RF field  $\mathcal{E}$  (with the fundamental frequency  $\omega_l$ ). The total polarization of the surface-state electrons could be simply described as  $\tilde{\mathcal{P}} = \mathcal{N}\mathcal{P}$ , with  $\mathcal{N}$  being the number of the electrons. Due to such a strong polarization  $\tilde{\mathcal{P}}$ , the frequency-doubled radiations  $\mathcal{E}_2$  (with frequency  $2\omega_l$ ) could be significantly generated. Liking a number of previous experiments [2, 3, 6], here the densities of electrons gas are set sufficiently low, such that the Coulomb interaction between the electrons does not affect significantly the expected

results. Furthermore, for achieving a considerable yield and a high directionality of the output lights, a resonant cavity is introduced in the setup liking that in the usual lasers. Experimentally, the transition frequency  $\omega_e$  between the ground and excited states of the surface-state electrons could be *linearly Stark-tuned* [2], typically about  $0.8 \text{ GHz (V/cm)}^{-1}$ , by applying the vertical electric fields. Such a linear Stark operation provides a large regime to set the work frequencies of electrons, e.g.,  $0.1 \sim 1 \text{ THz}$  or more. This means that the frequency of the produced laser of the surface-state electrons is adjustable, rather than that in the usual laser where the frequency of the produced laser is fixed (once the working materials are definitely selected).

In conclusion, we have proposed a new candidate: surface-state electrons on the liquid Helium, to realize the frequency-doubling radiations at high-frequency microwave regime. We have shown that, due to the broken parity-symmetries of the electronic surface-states, the like-Rabi oscillations could be effectively excited under the large-detuning drivings, and consequently the frequency-doubling radiations are generated. Remarkably, the proposed generation of frequency-doubling process could be immediately realized by the experimentally-existing setups. Interestingly, the present system could be utilized to realize the unusual frequency-doubling in THz regime, as the energy-splittings between the Rydberg states (e.g., the ground and first excited states) of the surface-state electrons are really at the order of THz.

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